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Selected Energy Epitaxial Deposition and Low Energy Electron Microscopy of AlN, GaN and SiC Thin Films

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13. ABSTRACT ***(Maximum 200 words)***

A study of the homoepitaxial growth of GaN(0001) layers was conducted *in situ* and in real time using the low-energy electron microscope. An evaporative cell supplied the Ga flux while the NH_3 flux was supplied via a seeded-beam supersonic jet source. At growth temperatures of 665° and 677°C, smooth GaN(0001) layers with well-defined step structures were grown on MOCVD-GaN(0001) substrates. In general, non-faceted homoepitaxial layers were achieved when the Ga/NH_3 flux ratios exceeded 2, starting with a Ga-covered substrate surface, in the temperature range 655-710°C. Preliminary kinetics data for homoepitaxial GaN films grown under Ga-stable conditions are consistent with an energy barrier for direct dissociative chemisorption of NH_3 on GaN(0001) of approximately 0.5 eV.

14. SUBJECT TERMS
GaN, homoepitaxial growth, low-energy electron microscope, LEEM, seeded-beam supersonic jet, flux ratios, screw dislocations, NH_3-seeded supersonic molecular beam, atomic force microscopy, growth rate, kinetic energy, surface morphology, cleaning

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I. Introduction

The realized and potential electronic applications of AlN, GaN and SiC are well known. Moreover, a continuous range of solid solutions and pseudomorphic heterostructures of controlled periodicities and tunable band gaps from 2.3 eV (3C-SiC) to 6.3 eV (AlN) have been produced at North Carolina State University (NCSU) and elsewhere in the GaN-AlN and AlN-SiC systems. The wide band gaps of these materials and their strong atomic bonding have allowed the fabrication of high-power, high-frequency and high-temperature devices. However, the high vapor pressures of N and Si in the nitrides and SiC, respectively, force the use of low deposition temperatures with resultant inefficient chemisorption and reduced surface diffusion rates. The use of these low temperatures also increases the probability of the uncontrolled introduction of impurities as well as point, line and planar defects which are likely to be electrically active. An effective method must be found to routinely produce intrinsic epitaxial films of AlN, GaN and SiC having low defect densities.

Recently, Ceyer [1, 2] has demonstrated that the barrier to dissociative chemisorption of a reactant upon collision with a surface can be overcome by the translational energy of the incident molecule. Ceyer's explanation for this process is based upon a potential energy diagram (Fig. 1) similar to that given by classical transition-state theory (or activated-complex theory) in chemical kinetics. The dotted and dashed lines in Fig. 1 show, respectively, the potential wells for molecular physisorption and dissociative chemisorption onto the surface. In general, there will be an energy barrier to overcome for the atoms of the physisorbed molecule to dissociate and chemically bond to the surface. Depending upon the equilibrium positions and well depths of the physisorbed and chemisorbed states, the energy of the transition state E^* can be less than zero or greater than zero. In the former case, the reaction proceeds spontaneously. In the latter case, the molecule will never proceed from the physisorbed state (the precursor state) to the chemisorbed state unless an additional source of energy can be drawn upon to surmount the barrier. This energy can only come from either (1) the thermal energy of the surface, (2) stored internal energy (rotational and vibrational) of the molecule, or (3) the incident translational kinetic energy of the molecule. Conversion of translational kinetic energy into the required potential energy is the most efficient of these processes. Moreover, by adjusting the kinetic energy, E_i, of the incoming molecule, it is possible to turn off the reaction ($E_i < E^*$), to tailor the reaction to just proceed ($E_i = E^*$), or to set the amount of excess energy to be released ($E_i > E^*$). The thrust of the present research is to employ these attributes of the beam translational energy to tune the reaction chemistry for wide band gap semiconductor epitaxial growth.

The transition state, E^*, is essentially the activation energy for dissociation and chemisorption of the incident molecules. Its exact magnitude is unknown, but is most certainly

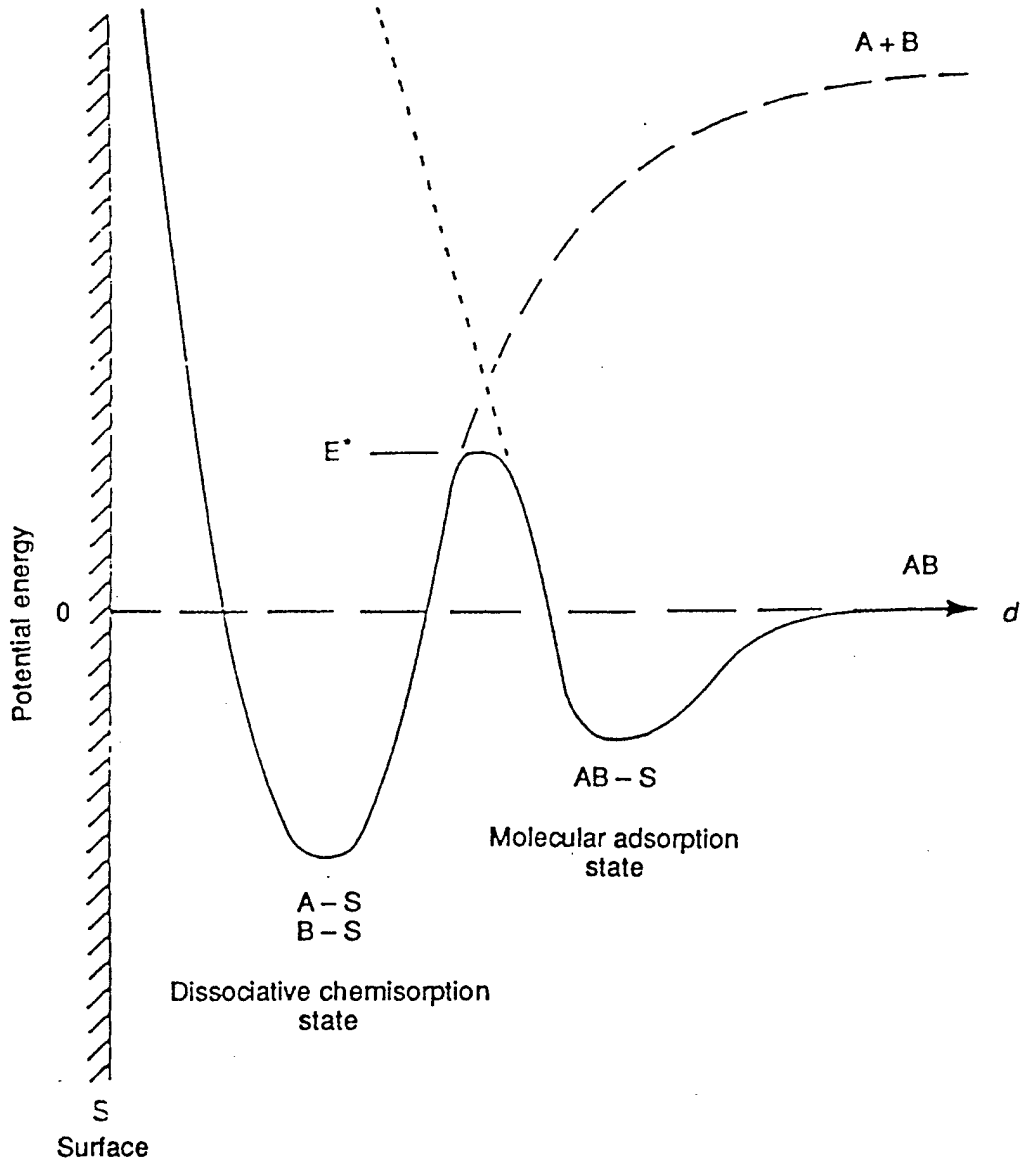

Figure 1. Schematic potential energy diagram of an activated surface reaction involving a molecularly physisorbed precursor state [from Ref. 1].

lower than the dissociation energy of the free molecule. It does not necessarily follow, however, that any kinetic energy above E^* will promote high-quality epitaxial growth of GaN. One must take into consideration another energy threshold, E_d, beyond which the kinetic energy of the incident flux will cause damage to the epitaxial film being synthesized. A typical Ed threshold value is approximately five times the band gap of the crystal and in the case of GaN, Ed ≈18 eV.

From the above consideration, it is clear that the key to high quality epitaxial growth is to be able to tune the energy of the incoming flux species over a range of energies defined by the window between E^* and E_d. Since the window is quite restrictive, i.e. 1-20 eV, it is essential that the energy spread of the flux species must be small, i.e. the flux species should ideally be

monoenergetic. To this end, we employ selected energy epitaxial deposition (SEED) systems for the growth of AlN, GaN and SiC wide band gap semiconductors. The SEED systems are of two types: (1) a seeded-beam supersonic free-jet (SSJ) and (2) a dual ion-beam Colutron. Both these SEED systems have the desirable property of a narrow energy spread of ≤ 1 eV.

Epitaxial growth using the seeded-beam SSJ involves a close collaboration between investigators at NCSU and Arizona State University (ASU). At ASU, the SSJ is interfaced directly into a low-energy electron microscope (LEEM) for the conduct of *in situ* studies of the nucleation and growth of epitaxial layers; while at NCSU, the SSJ systems are used to grow device-quality AlN, GaN and SiC for real applications. Exchanges in personnel (students) and information between the two groups ensures the achievement of desired results. The additional thin film growth experiments using dual-beam colutrons and the theoretical studies referred to in this report are primarily conducted at ASU.

The research conducted in this reporting period and described in the following sections has been concerned with (1) an *in situ* study in real time of the homoepitaxial growth of GaN(0001) using the low-energy electron microscope, and 2) investigation of the homoepitaxial growth of GaN films under Ga-stable conditions consistent with an energy barrier for direct dissociative chemisorption of NH_3 on GaN(0001) of approximately 0.5 eV. The following individual sections detail the procedures, results, discussions of these results, conclusions and plans for future research. Each subsection is self-contained with its own figures, tables and references.

1. S. T. Ceyer, Langmuir 6, 82 (1990).
2. S. T. Ceyer, Science 249, 133 (1990).

II. Low-energy Electron Microscopy Observations of GaN Homoepitaxy Using a Supersonic Jet Source

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A study of the homoepitaxial growth of GaN(0001) layers was conducted *in situ* and in real time using the low-energy electron microscope. The Ga flux was supplied by an evaporative cell while the NH_3 flux was supplied via a seeded-beam supersonic jet source. At growth temperatures of 665° and 677°C, smooth GaN(0001) layers with well-defined step structures were grown on MOCVD–GaN(0001) substrates. In general, non-faceted homoepitaxial layers were achieved when the Ga/NH_3 flux ratios exceeded 2, starting with a Ga-covered substrate surface, in the temperature range 655–710°C.

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The selected energy epitaxy (SEE) approach of GaN growth is based on a concept demonstrated by Ceyer [1, 2] that upon collision with a surface, a reactant can overcome the potential barrier to dissociative chemisorption by its translational energy. In this case, the reactant molecules, NH_3, are seeded into a beam of the He atoms from a supersonic jet (SSJ) source. We have recently shown that with the SSJ, essentially monoenergetic beams of NH_3 in the energy range of 0.20–0.65 eV with a FWHM of 0.1 eV can be easily produced [3-5]. Such an approach has the advantage of fine-tuning an energy window in which dissociation and reaction are promoted, surface diffusion is enhanced, while defect creation due to energetic collisions is minimized. All of these processes can, in principle, take place at a lower growth temperature since part of the translational energy of the flux species is converted into potential energy to overcome the activation barrier while the rest goes to promote mobility of the absorbed species to migrate to preferred sites such as step edges on the surface. Since the activation barrier for dissociation and chemisorption of NH_3 on a GaN(0001) surface has been calculated to have an upper limit of 0.5 eV [6] and experimentally determined to be 0.25±0.1 eV [5], the SSJ is ideally suited for low-temperature GaN growth. A review of epitaxy of Group III nitrides by SSJ has been given recently by Ferguson and Mullins [7]. In this report, we describe *in situ* real-time observations of GaN homoepitaxy by SSJ using a low-energy electron microscope (LEEM) [8]. The experimental parameters, i.e. substrate preparation, growth temperature and flux ratio, leading to smooth basal plane (0001) growth were explored and determined in our LEEM studies.

The experimental configuration for conducting the LEEM observations of GaN growth by SSJ has been described in detail previously [4]. The base pressure in the LEEM was ~1×10^{-10} Torr, which increased to 3×10^{-8} Torr when the SSJ was in operation. The typical flux rates were $(1–10)\times10^{13}$ cm^{-2} s^{-1} for Ga from an evaporative cell, and $(0.3–3)\times10^{13}$ cm^{-2} s^{-1} NH_3 from the SSJ. Thermal desorption measurements by Ambacher *et al.* [9] show that the onset of GaN decomposition occurs at 750°C, a result supported by the maximum deposition rate occurring at 760–780°C observed by Lee *et al.* [10] in gas source molecular beam epitaxy (GSMBE) of GaN(0001) with fluxes of 6×10^{14} cm^{-2} s^{-1} for Ga and 1×10^{17} cm^{-2} s^{-1} for NH_3. Because of the low growth rates in the LEEM, in order to avoid decomposition, we chose to conduct the growth experiments in the temperature range of 665–710°C.

The substrates used in the present work were 1.4 μm thick GaN(0001) layers grown by MOCVD on 0.1 μm thick AlN buffer layers on 6H-SiC(0001) mounted on a rotating susceptor described previously [11]. The substrate surface was cleaned in the LEEM by exposure to a flux of N-atoms from an EPI RF-plasma source operated at 200 W and a nitrogen pressure of ~1×10^{-5} Torr for 10–15 min around 675°C. After the cleaning procedure,

the GaN substrate surface usually displayed a ($\sqrt{3}$ x $\sqrt{3}$) LEED pattern. Additional treatment by Ga deposition in the temperature range of 600–730°C led to a (2×2) LEED pattern on some of the substrates.

Figure 1 shows frame-captured LEEM images of GaN homoepitaxial growth at 677°C with the NH_3 flux at 0.7×10^{13} cm^{-2} s^{-1} and Ga flux at 5.9×10^{13} cm^{-2} s^{-1}, i.e. a Ga/NH_3 flux ratio of ~8. After 6 min. of deposition, the meandering steps of the substrate surface in frame (a) were largely obscured, replaced by a grainy surface morphology in frame (b). After 9 min. of deposition, the grainy appearance was still present, but the meandering step structure began to reemerge in frame (c). After about 2 hours of deposition, the meandering steps were restored and the surface showed graininess too fine to be seen in frame (d), but clearly visible in the original images. The GaN film surface did not change significantly after frame (d),

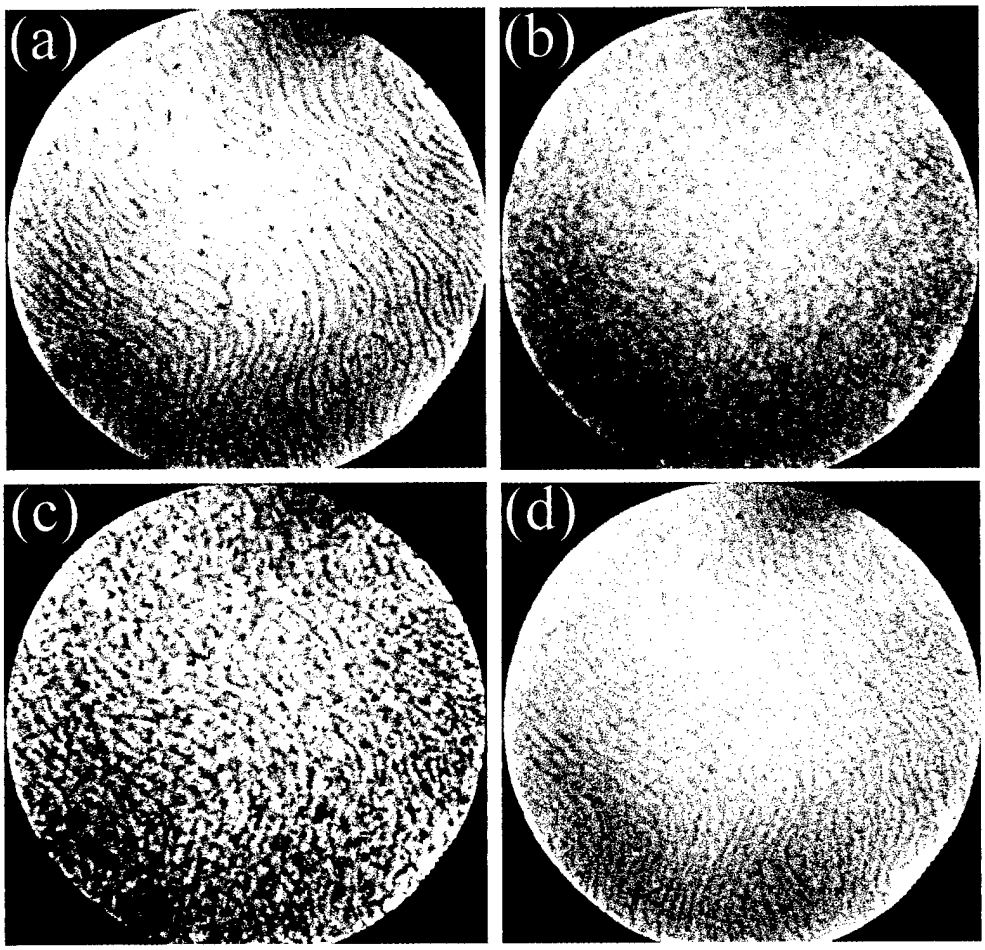

Figure 1. Frame-captured LEEM video images of homoepitaxial growth of GaN on a MOCVD GaN(0001) substrate: (a) initial ($\sqrt{3}$ x $\sqrt{3}$) + (2×2) surface of the GaN substrate; (b) after 6 min. of deposition; (c) after 9 min.; and (d) after 120 min. Substrate temperature 677°C. Ga flux 5.92×10^{13} cm^{-2} s^{-1}; NH_3 flux 0.74×10^{13} cm^{-2} s^{-1}. Electron energy 11.0 eV. Field of view 4.8 μm.

retaining the same appearance even after 2 more hours of deposition. The LEED pattern of the surface throughout this growth sequence was (5×5) after 20 min., then became (1×1) after 40 min. No facet spots were observed.

The frame-captured LEEM images shown in Fig. 2 were obtained on a different substrate with the same flux ratio of ~8 but at half the flux rates at a growth temperature of 665°C. The surface morphology during growth followed an almost identical sequence as that shown in Fig. 1. After 30 min. of deposition, the surface appeared grainy in frame (b) and the meandering step structure of the substrate in frame (a) was completely masked by the graininess. The meandering steps reemerged in frame (c), after 70 min. of deposition. The step structure was restored and sharpened after further deposition as shown in frame (d) taken

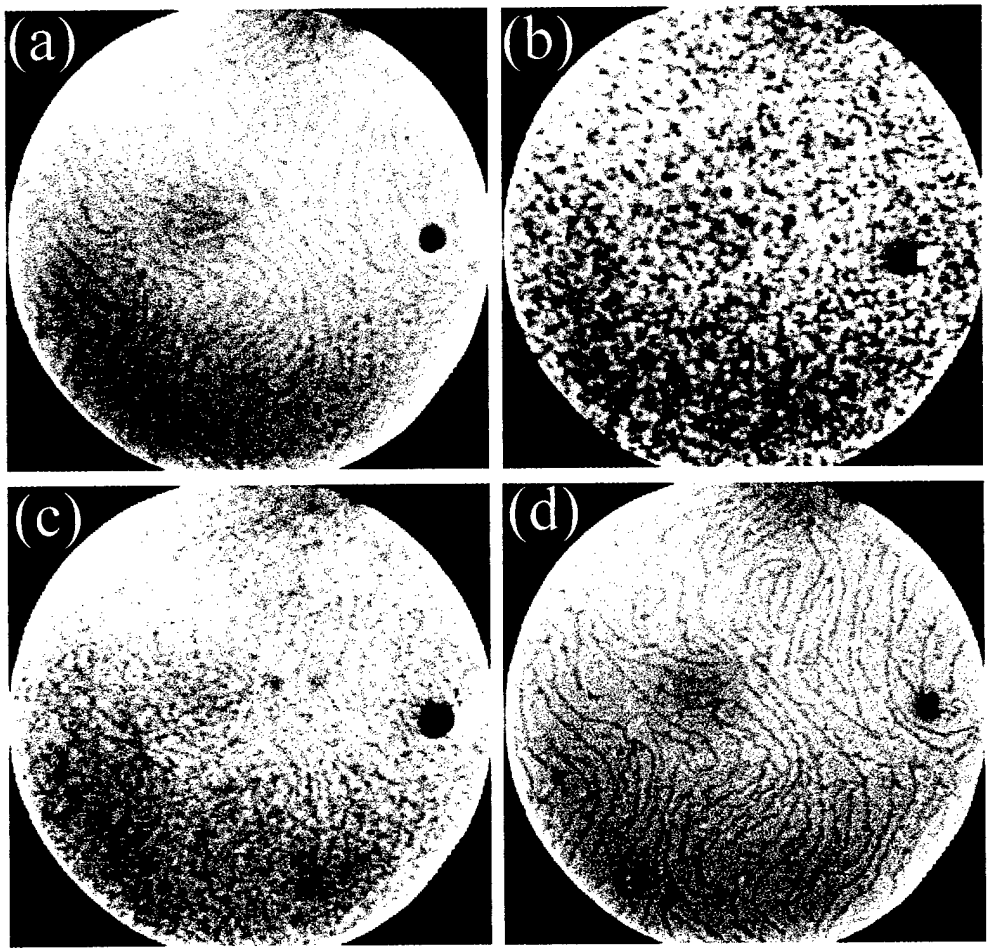

Figure 2. Frame-captured LEEM video images of homoepitaxial growth of GaN on a MOCVD GaN(0001) substrate: (a) initial ($\sqrt{3}$ x $\sqrt{3}$) surface of the GaN substrate; (b) after 30 min. of deposition; (c) after 70 min.; and (d) after 210 min. Substrate temperature 665°C. Ga/NH_3 flux ratio ~8 as in Fig. 1, but the individual flux rates were halved. Electron energy 10.8 eV. Field of view 4.8 μm.

at 210 min. Further deposition for 3 hours did not produce significant change in surface morphology. The LEED pattern of the surface was (2×2) after 13 min. and then (1×1) after 57 min. The (1×1) pattern remained throughout growth without facet spots indicating (0001) basal plane growth. We have thus shown that under similar growth conditions, i.e. a Ga/NH_3 flux ratio of ~8 and growth temperatures in the range of 665–677°C, basal plane growth of GaN(0001) can be achieved. Figure 2 shows slower growth because the individual Ga and NH_3 flux rates used were half of those used in obtaining the growth sequence in Fig. 1.

We interpret our *in situ* real-time LEEM/LEED observations of GaN homoepitaxy as quasi-two-dimensional island growth similar to the model proposed by Headrick *et al.* [12] for GSMBE of cubic GaN of β-SiC(001). Frames (b) of both Figs. 1 and 2 represent the initial growth of islands large enough to be resolved by the LEEM. The lateral growth of these islands led to a continuous layer. Once this layer was formed, the meandering steps in frames (d) resumed the sharper appearance as those shown in frames (a) of Figs. 1 and 2. Growth continued at a steady rate in islands at the resolution limit of our LEEM. Hence, frames (d) in both Figs. 1 and 2 appear unchanged over two or more hours of deposition. This conjecture is supported by *ex situ* atomic force microscopy (AFM) images taken on the GaN homoepitaxial layer shown in Fig. 1(d). The AFM image in Fig. 3 shows that the terraces consisted of ~10 nm sized grains or islands, in qualitative agreement with the resolution limit of our LEEM. The nucleation of these small islands with (0001) basal plane as indicated by the (1×1) LEED pattern with a strong (00) spot is probably due to a change in

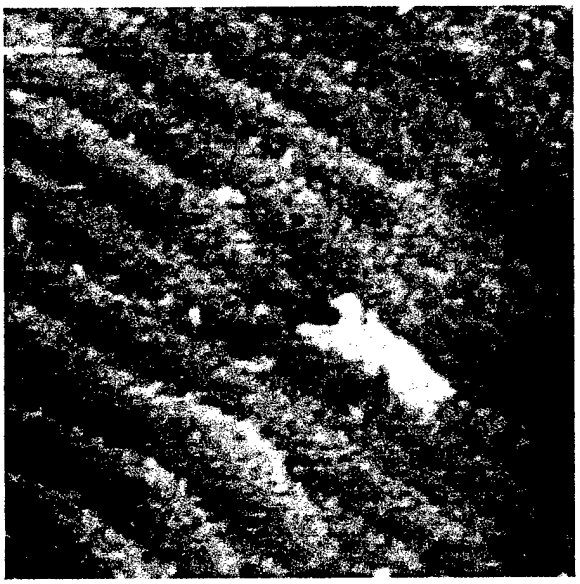

Figure 3. AFM image of the GaN(0001) homoepitaxial layer as shown in frame (d) of Fig. 1. Scan area 1 μm × 1 μm.

the growth kinetics. The initial nucleation occurred on a chemically modified MOCVD GaN(0001) substrate surface possibly not completely cleaned even after our cleaning treatment. The initially completed homoepitaxial GaN(0001) layer was of higher purity because it was grown under UHV conditions. The nucleation rate on this pure surface was much higher leading to a high density of very small 2D crystals which coalesced before they could be resolved in LEEM. Thus, while film growth continued after frame (d), the grainy appearance in frames (b) and (c) of the LEEM images never reappeared.

Our *in situ* real-time LEEM investigations of GaN homoepitaxy by SSJ leading to basal plane growth are summarized in Fig. 4 where we have plotted the Ga/NH_3 flux ratio versus the growth temperature in the range of 665–710°C. The different symbols in Fig. 4 indicate the LEED patterns observed for the substrates after the N-atom cleaning and Ga deposition treatment described earlier. It appears that as long as the Ga/NH_3 flux ratio was greater or equal to 2, non-faceted growth was achieved. We should point out that the term "non-faceted" growth refers to all forms of (0001) basal plane growth which do not contain facets, i.e. the LEED pattern shows (1×1) only with a strong (00) LEED spot. However, not all non-faceted growth exhibited a well-defined step structure such as those shown in frames (d) of Figs. 1 and 2. Some of the non-faceted layers show a grainy appearance in the LEEM images. AFM measurements of these surfaces do not produce any meaningful step heights between two adjacent grains. When the Ga/NH_3 ratio falls below 2, facet spots appear in the LEED pattern, the (00) spot fades in intensity and the LEEM image shows a grainy and spotty surface. While we found that we could always convert a non-faceted GaN layer into a faceted layer simply by lowering the Ga/NH_3 flux ratio to <2, we could not convert a well-developed faceted layer back into basal plane growth by increasing the flux ratio.

In summary, we have achieved smooth basal plane layer growth in GaN(0001) homoepitaxy using a NH_3 seeded-beam SSJ at a growth temperature range of 665–710°C. *In situ* real-time LEEM combined with LEED is a powerful method for the study of growth processes because the instant feedback it provides allows changing of parameters to optimize growth during observation.

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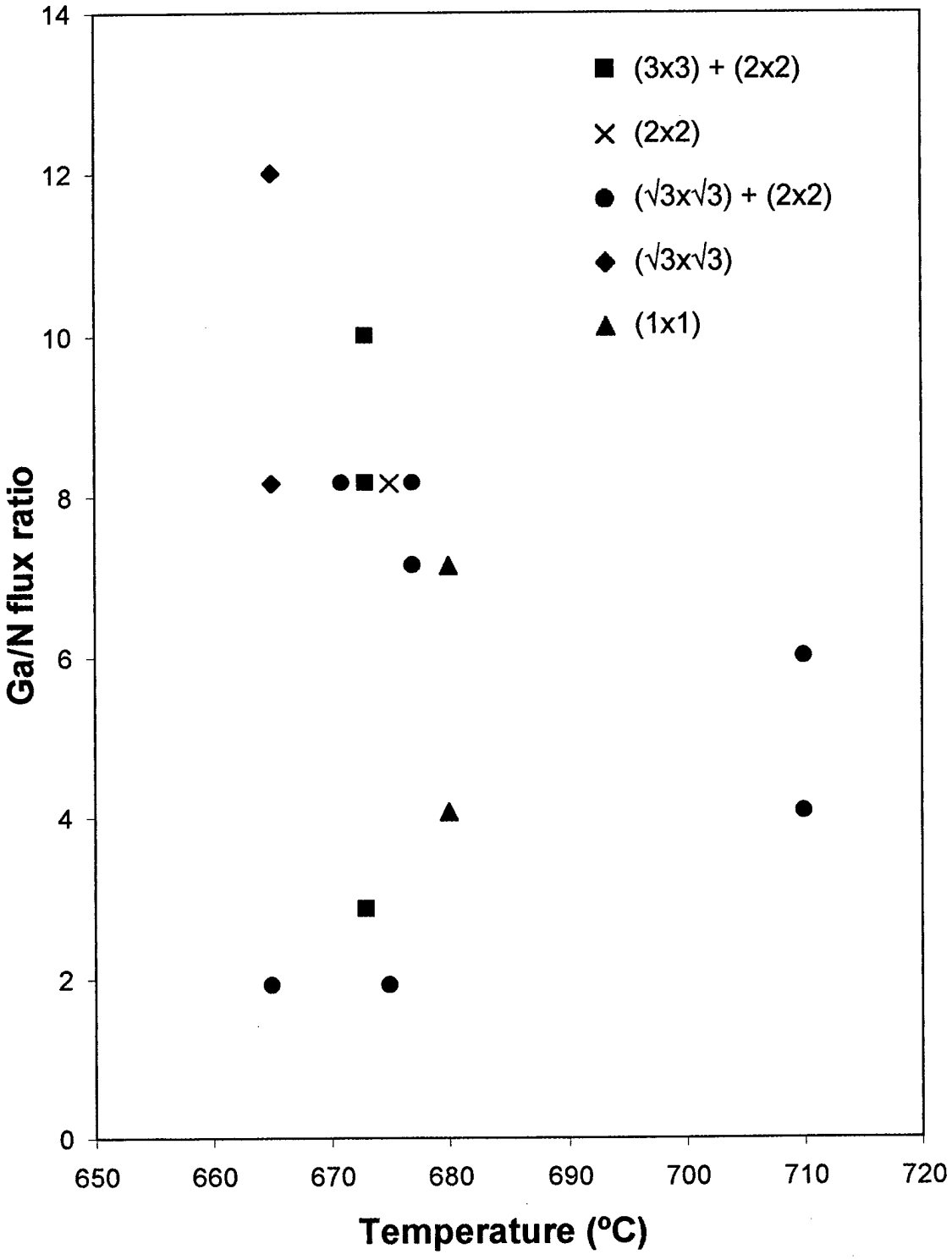

Figure 4. A plot of Ga/NH_3 flux ratio versus growth temperature illustrating non-faceted homoepitaxial growth. The ranges of flux rates are: (1–10) × 10^{13} cm^{-2} s^{-1} for Ga, and (0.3–3) × 10^{13} cm^{-2} s^{-1} for NH_3. The different symbols indicate the LEED patterns observed for the substrate surfaces prior to growth.

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II. Optimization of Homoepitaxial Growth of GaN Using NH_3-seeded Supersonic Beams

A. Introduction

Selected energy epitaxy (SEE) using NH_3-seeded supersonic beams is an alternative to plasma-source molecular beam epitaxy (PSMBE) for low-temperature growth of III-nitride semiconductors. We have chosen to focus on homoepitaxial growth of GaN thin films employing MOCVD-grown GaN/SiC substrates. In the December 1998 report, we demonstrated that smooth homoepitaxial GaN films can be grown by SEE using NH_3-seeded supersonic beams under Ga-rich conditions. Preliminary data reported herein are consistent with an energy barrier for NH_3 dissociative chemisorption of approximately 0.5 eV. Unfortunately, the films contained unacceptably high carbon and oxygen concentrations (10^{18} cm^{-2} each). Several sources of contamination were identified, and corrective action was taken through a series of equipment modifications. After the modifications, a GaN sample grown by conventional gas-source MBE using an NH_3 leak evidenced dramatically lower carbon and oxygen contamination levels.

B. Experiment

The substrates were 1-2 μm thick GaN films grown by MOCVD at 1050°C on 6H-SiC using a 0.1 μm AlN buffer layer. Wafers of GaN/AlN/SiC were obtained from Prof. Davis' group. A tungsten film (~0.1 μm) was deposited on the SiC face, and approximately 1×1 cm^2 samples were cut using a diamond saw. Samples were degreased using trichloroethylene (TCE) prior to mounting on a Mo sample holder.

The SEE growth system has been described in detail in previous reports. Recent equipment modifications include: a new Mo sample holder that utilizes direct radiative heating, a liquid-nitrogen cooled cryopanel for pumping condensable vapors during cleaning and deposition, and new bakeout bands for the growth chamber. With the new sample holder, backside metallization of the GaN/AlN/SiC substrates is required (see above), and these changes also necessitated recalibration of the optical pyrometer. The major advantage of this heating arrangement is that it obviates the need for Ag paste (formerly used to provide thermal contact between the sample and the Mo block), thus eliminating a potential source of contamination. The cyropanel handles most of the NH_3 pumping load during deposition and also reduces the partial pressure of H_2O, minimizing this potential source of oxygen contamination. The base pressure of the growth chamber was reduced from 2×10^{-8} to 1×10^{-9} Torr by using new Watlow bakeout bands on the larger ports.

The Ga flux to the substrate was measured using a water-cooled quartz crystal monitor. The monitor was placed in the sample position facing the Knudsen cell. The measured flux,

assuming a Ga sticking coefficient of unity, was correlated with the K-cell temperature (950-1020°C) using the following relationship:

$$\ln(F_{Ga}) = 55.29 - \frac{26550}{T}$$

where F_{Ga} is the Ga flux in $cm^{-2}\ s^{-1}$ and T is the absolute temperature in K. The actual flux to the substrate during growth will be only 50% of this value, owing to the 60° angle between the K-cell and the substrate.

C. Results and Discussion

The growth kinetics for experiments using supersonic beams of 10% NH_3 seeded in He are presented in Fig. 1. The growth rate at 700°C has been converted to an equivalent GaN flux (F_{GaN}) using the following relationship:

$$F_{GaN} = \frac{\rho_{GaN}(G)N_{Av.}}{M_{GaN}}$$

where ρ_{GaN} is the GaN density, G is the growth rate, $N_{Av.}$ is Avogardro's number, and M_{GaN} is the formula weight of GaN. Figure 1 illustrates that the growth rate increases almost linearly with F_{Ga}, indicating Ga-limited (N-stable) growth under these conditions. Moreover, the data evidence that the 10% NH_3-seeded supersonic beam provides an active nitrogen flux of $>4\times10^{14}\ cm^{-2}\ s^{-1}$. The 45° line in the Fig. 1 corresponds to 100% Ga incorporation. The actual Ga incorporation efficiency decreases from 100% to approximately 90% as the incident Ga flux is increased. The lowest Ga incorporation efficiency (82%) was observed for films grown using 10% NH_3 and a 600°C nozzle temperature (0.61 eV kinetic energy) with an incident Ga flux of $3.2\times10^{14}\ cm^{-2}\ s^{-1}$. We infer that under N-stable conditions increasing NH_3 kinetic energy (in the limited range of 0.33-0.61 eV) tends to either have no effect or a small negative effect of the growth rate.

Preliminary experiments were performed to examine the effects of NH_3 kinetic energy on growth kinetics in the N-limited (Ga-stable) growth regime. Seeded supersonic beams containing 3% NH_3 in He or in Ar were used, and 2-h growth runs were conducted at 700°C using Ga K-cell temperatures of 980° and 990°C. The results are given in Table I.

The observed growth rates using 0.48 and 0.74 eV NH_3 beams are nearly equal; however, the growth rate for the 0.05 eV NH_3 beam was negligibly small. The data indicate that there is a potential energy barrier (E_B) for direct dissociative chemisorption of NH_3 on GaN. An accurate determination of the barrier height cannot be made from these data, but the barrier is in the range: $0.05<E_B<0.48$ eV. The data are consistent with those reported by our colleagues at ASU in the December 1997 Quarterly Progress Report. The ASU group has also reported theoretical calculations in which the chemisorption barrier was estimated at 0.5 eV.

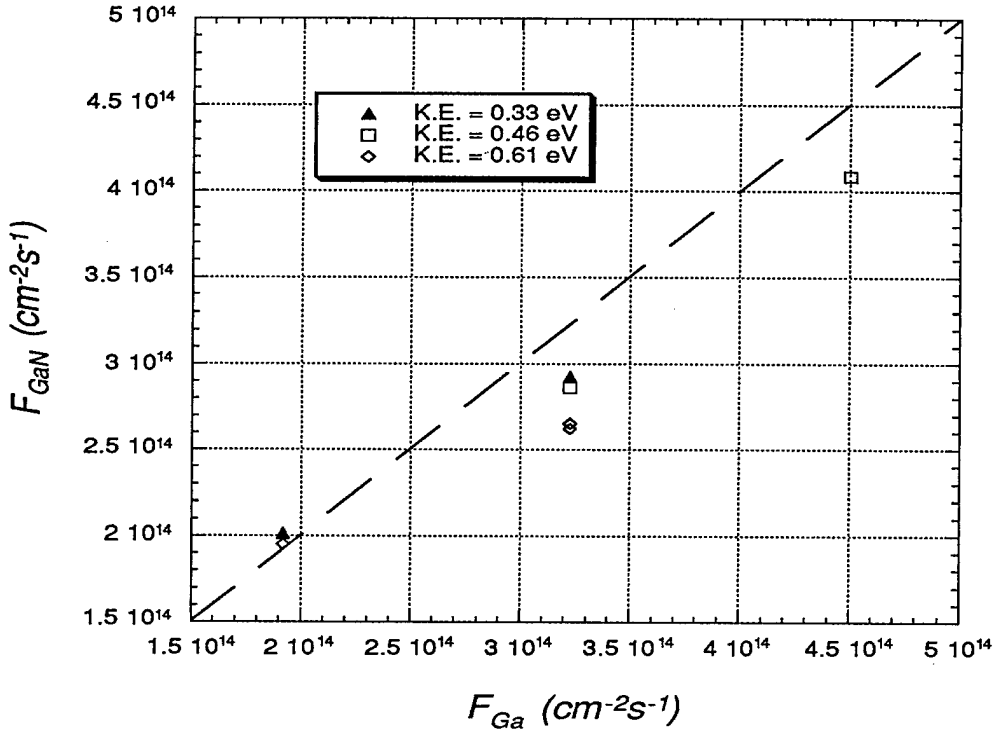

Figure 1. GaN growth kinetics under N-stable conditions. Substrate temperature = 700°C.

Table I. Effect of NH_3 Kinetic Energy on GaN Growth Rate in the Ga-stable Regime

Ga Flux (cm^{-2} s^{-1})	Bath Gas	NH_3 K.E. (eV)	Growth Rate (nm/h)
3.8×10^{14}	He	0.74	230
3.8×10^{14}	He	0.48	215
3.8×10^{14}	Ar	0.05	0

The GaN films deposited in these experiments contained unacceptably high levels of carbon and oxygen contamination (10^{18} cm^{-2} for each element), as determined by secondary ion mass spectroscopy (SIMS). Moreover, x-ray photoelectron spectroscopy (XPS) of the surfaces of the deposited films evidenced contamination levels of 2-5 at% carbon and oxygen. Investigation revealed that the contamination originated from inadequate bakeout of the growth chamber and the use of contaminated sample holders. Residual gas analysis (RGA) after a

standard 24-h bakeout evidenced that H_2O and CO were the primary background constituents. H_2O accounted for most of the 2×10^{-8} Torr base pressure, indicating that the chamber was leak-tight but that the bakeout temperature was too low. New band heaters were installed on the larger ports; silicon-rubber heating patches were applied to the chamber body; and a new bakeout procedure was implemented requiring that the chamber be baked at 150°C until the pressure falls below 2×10^{-7} Torr. These changes reduced the base pressure by approximately one order of magnitude to the low 10^{-9} Torr range. The old solid Mo sample holders were another source of contamination. Buildup of solvent residues from the colloidal Ag paste which was used to mount the samples was the main problem. Re-design of the sample holder for direct radiative sample heating eliminated this contamination source.

A GaN control sample was grown in the modified SEE system by conventional gas-source MBE at 700°C. NH_3 was leaked into the growth chamber to establish a background pressure of 1×10^{-5} Torr. XPS analysis of the substrate after cleaning in an NH_3 ambient at 815°C evidenced a negligible oxygen concentration and <1 at% carbon. Analysis by XPS of the GaN film gave similarly encouraging results: <1 at% carbon and oxygen. Analysis by SIMS of the film is planned to confirm the anticipated reduction in carbon and oxygen contamination.

D. Future Plans

Future studies include the investigation of NH_3 decomposition on the Ga-polar GaN(0001) surface as a function of substrate temperature and NH_3 incident translational energy using *in situ* mass spectroscopy; growth and characterization of GaN films under Ga-stable conditions using NH_3-seeded supersonic beams; and, investigation of the effects of translational energy on the decomposition of triethlygallium (TEG) on GaN(0001) surfaces using XPS and desorption mass spectroscopy.

IV. Distribution List

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